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Microplastics in an urban wastewater treatment plant: The influence of physicochemical parameters and environmental factors

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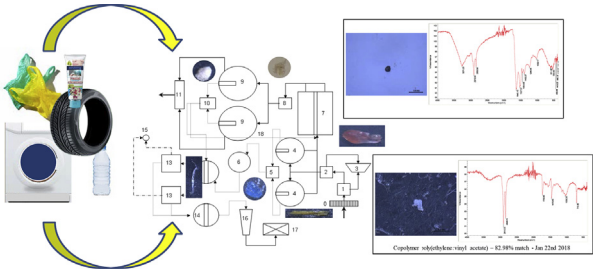
HIGHLIGHTS

- A significant removal of microlitter and microplastics through the WWTP.
- Biodegradable polymers proved to disappear in the final effluent.
- High suspended solids were correlated to low MP burden and an increasing size.
- No seasonal variability in the effluent, despite an increase during the warm season.

GRAPHICAL ABSTRACT



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ABSTRACT

This paper presents the abundance, concentration and variability of microplastics (MP) in an urban wastewater treatment plant (WWTP), according to different water parameters and environmental factors, their possible sources and removal efficiency. A total of 352.6 L of wastewater from four stages of the treatment process were processed following a standardized extraction protocol by density separation, trinocular microscopic identification and polymeric analysis by Fourier transform infrared spectroscopy. MP comprised a 46.6% of total microlitter, with a statistically significant removal of 90.3% in the final effluent of the WWTP. Five different shapes were isolated; i.e. fragment, film, bead, fiber, and foam. The most prominent MP forms in the final effluent were fragments and fibers, with the most common size class being 400–600 μm. Seventeen different polymer families were identified, with low-density polyethylene being the most prevalent one (52.4%) in a film form (27.7%), mostly from agriculture greenhouses near the sewage plant and single plastic bags (it is noted that only a year ago consumers are charged for them in Spain). Influent wastewater with high concentrations of suspended solids proved to have a low MP burden with a larger MP size, possibly due to a hetero-aggregation with particulate matter. Agglomeration of polystyrene and polyethylene terephthalate with organic material is also suggested, both with surface energies higher than 25 mN m^{-1} enough for a high biofouling rate. The sewage plant cushions sharp-point microplastic concentrations during the warm season, allowing a stable performance of the WWTP.

1. Introduction

The figures of annual plastic production in Europe have been reported to grow from 47.8 million tonnes in 2014 up to 60 million tonnes in 2016, with a 3.3% increase compared to 2015

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(PlasticsEurope, 2017). Microplastics (MP), as an emerging pollutant detected in the environment with a size below 5 mm (Lambert and Wagner, 2016), can be originated by weathering, photolysis, abrasion, mechanical and microbial decomposition of discarded macroplastics in the environment (Ziajahromi et al., 2017), making up secondary MP that long outlast the consumers who used them. In contrast, primary MP are intentionally manufactured with a microscopic size (Estahbanati and Fahrenfeld, 2016); i.e., preproduction pellets, plastic granulates, air blasting and microbeads for personal care products (Bayo et al., 2017).

Since the term was first identified by Carpenter and Smith (1972) as a pollutant in the Sargasso Sea, MP pollution has been of global concern in marine ecosystems, with much less information in freshwater environments. Recent studies of MP accumulation in water of lakes (Eriksen et al., 2013; Ballent et al., 2016), rivers (Besseling et al., 2017; Wang et al., 2017), Arctic ice (Obbard et al., 2014; La Daana et al., 2018), lagoons (Vianello et al., 2013; Abidli et al., 2017), runoff (Andrady, 2011; Duis and Coors, 2016), estuaries (Bakir et al., 2014; Gallagher et al., 2016) and beaches (Liebezeit and Dubaish, 2012; Wright et al., 2013; Laglbauer et al., 2014) have proven to be a major pathway to reach the ocean (Eerkes-Medrano et al., 2015). Besides, MP generated from fishing nets and directly emitted into the seas (Dümichen et al., 2017). Different marine species have been reported to ingest MP, possessing varied feeding strategies and occupying different trophic levels (Eerkes-Medrano et al., 2015). This ingestion may cause blockage of the digestive tract, reduce food consumption due to false feelings of satiety, internal wounds and death (Gallagher et al., 2016). Furthermore, the sorptive function of MP for both inorganic and organic pollutants has been widely studied (Gauquie et al., 2015; Wang et al., 2015; Bayo et al., 2017), acting as a carrier to concentrate and transfer all these pollutants to different organisms, despite their desorption is likely to be slower than from dissolved organic matter and it is governed by its content (Zuo et al., 2019).

Since Browne et al. (2011) reported polyester and acrylic fibers in shoreline sediment samples similar to that collected from wastewater treatment plants (WWTPs) effluents, further studies have addressed this land-based pathway as a sink and source of MP, investigating the abundance, morphology, color, and characterization of these microparticles (Talvitie et al., 2015; Estahbanati and Fahrenfeld, 2016; Ziajahromi et al., 2017) with varied results. Talvitie et al. (2015) reported a 25 times higher concentration of fibers in the effluent of a WWTP compared to the receiving sea, while Murphy et al. (2016) reported that 65 million MP could enter the aquatic environment every day from a sewage plant located in Scotland, despite a 98.41% removal rate. Concerning methodology, Ziajahromi et al. (2017) validated a method to sample and process microplastics from 3 WWTPs in Sidney, Australia, and Kalčíková et al. (2017) have developed a lab-scale batch reactor to confirm a 52% average capture of microbeads by activated sludge.

WWTPs are complex systems with chemical, physical and biological processes taking place simultaneously, in order to achieve a high-quality final effluent to be reused in agriculture or returned to the environment. Although WWTP effluent has been recognized as an important source of MP to the aquatic environment (Roex et al., 2013), some additional information still remains unknown, as the role of physicochemical and environmental factors in the behavior of MP. In this paper we report the abundance, concentration and variability of microplastics according to different wastewater parameters and environmental factors, the matching between demanded polymers and microplastics isolated in wastewater samples, their possible sources, and the removal efficiency in a classical WWTP with activated sludge process (ASP).

2. Materials and methods

2.1. Sample collection and processing

Wastewater samples were collected at four stages of the treatment process (Table S1); i.e., grit and grease removal (GGR, 60.1 L), in order not to block or clog filters and pumps with large debris, primary clarifier (PCL, 59.3 L), activated sludge reactor (BRT, 103.4 L), and the effluent after the secondary clarifier (EFF, 143.0 L), from the urban WWTP "Cabezo Beaza", situated in Cartagena (Spain). This plant has been thoroughly described elsewhere (Bayo and López-Castellanos, 2016; Bayo et al., 2016), receiving wastewater from both urban and industrial activities. It consists of a conventional ASP with a primary treatment and two parallel activated sludge bioreactors, serving about 210,000 equivalent inhabitants (35,000 $m^3 d^{-1}$). A total of 125 grab samples were collected between September 2016 and April 2018, comprising 28 samples from GGR, 28 from PCL, 33 from BRT, and 36 from EFF. Sample volumes were precisely measured in each experiment, ranging from 0.5 to 3.7 L for GGR (average 2.1 ± 0.3 L), 0.5−3.6 L for PCL (average 2.1 ± 0.3 L), 1.4−3.7 L for BRT (average 3.1 ± 0.1 L), and 2.7−17.0 L for EFF (average 4.0 ± 0.4 L). Samples were always grabbed in glass bottles with metallic lid and, depending on the sampling point, volumes ranged from 0.47 to 13 L. They were acquired both in the morning (09:00−11:00, 197.0 L) and in the afternoon (15:00−17:00, 168.8 L), in order to determine any diurnal variation in the MP concentration.

Except for the EFF, that was directly filtered through the paper filter, all wastewater-based MPs were obtained by means of a density separation method with a salt-saturated solution of 120 g L^{-1} NaCl (2.05 M) (Panreac, Barcelona, Spain) and a final density of 1.08 g ml^{-1}, as previously reported (Bayo et al., 2019). The mixture, containing one parts salt solution and three parts of a wastewater sample, was placed in a 2 L glass beaker and mechanically stirred in a jar-test device for 20 min (300 rpm), which favors microplastic floatation. After 45 min of settlement, supernatant was vacuum filtered through a Büchner funnel using a paper filter (Prat Dumas, Couze-St-Front, France, diameter 110 mm, pore size 0.45 μm). The funnel wall was twice washed with bi-distilled water and also filtered. Isolated microlitter particles (ML) were recovered by orbital shaking (150 rpm, 30 min), placing filters into 120 mm glass Petri dishes after washing with 15 ml bi-distilled water. Samples were dried overnight at 100 °C in a forced air stove FD 23 (Binder GmbH, Tuttlingen, Germany). All experiments were carried out at room temperature (293 K).

To reduce the risk of contamination, only clothes made of natural fabric and clean cotton lab coats were worn by the analysts. The use of plastic lab devices was limited to the maximum, although it could not be entirely avoided. All glassware was thoroughly washed with tap water and twice with bi-distilled water after each experiment, covering it with aluminum foil to mitigate contamination.

Moreover, negative control samples or procedural blanks were analyzed throughout the study by vacuum filtering 1.5 L of bi-distilled water and 500 ml of salt solution through a clean paper filter, to determine any potential microplastic contamination during the lab work. Only one nylon and one cellulose fiber were detected in the blanks, and corrected in the corresponding samples.

2.2. Microplastic analysis and dataset

Posible microplastic particles were examined under an Olympus SZ-61TR Zoom Trinocular Microscope (Olympus Co., Tokyo, Japan) coupled to a Leica MC190 HD digital camera and an image capturing software Leica Application Suite (LAS) 4.8.0 (Leica Microsystems

Ltd., Heerbrugg, Switzerland), used for the analysis and recording of color, shape and size of each ML in its longest dimension. Once the images were captured, particles were successfully isolated in a 40-mm glass Petri dish for further analysis by Fourier transform infrared spectroscopy (FTIR).

FTIR was used for the identification of functional groups and molecular composition of polymeric surfaces. Samples were compressed in a diamond anvil compression cell, and spectra were acquired with a Thermo Nicolet 5700 Fourier transformed infrared spectrometer (Thermo Nicolet Analytical Instruments, Madison, WI, USA), provided with a deuterated triglycine sulfate, DTGS, detector and KBr optics. The spectra collected were an average of 20 scans with a resolution of 16 cm^{-1} in the range of 400–4000 cm^{-1}. Spectra were controlled and evaluated by the OMNIC software without further manipulations, and polymers were identified by means of different reference polymer libraries, containing spectra of all common polymers; i.e., Hummel Polymer and Additives (2011 spectra), Polymer Additives and Plasticizers (1799 spectra), Sprouse Scientific Systems Polymers by ATR Library (500 spectra), Rubber Compounding Materials (350 spectra), and literature (Hummel, 2002). In this sense, we followed the standard criteria reported by Frias et al. (2016), regarding a percentage match >60% between sample and reference spectra, with clear evidences of known peaks corresponding to different polymers (see Fig. S1).

The following water quality parameters were included in the dataset: water flow [WF] (m^3 d^{-1}), pH (pH units), water temperature [WT] (ºC), electrical conductivity [EC] (mS cm^{-1}), suspended solids [SS] (mg L^{-1}), chemical oxygen demand [COD] (mg L^{-1}), biochemical oxygen demand [BOD] (mg L^{-1}), ammonium nitrogen [NH_4–N] (mg L^{-1}), total nitrogen [TN] (mg L^{-1}) and phosphate phosphorus [PO_4–P] (mg L^{-1}). EC was determined with a Crison GLP 32 conductimeter (Barcelona, Spain) and pH with a Crison GLP 22 pH meter (Barcelona, Spain), both calibrated by means of standard solutions. COD was determined using the Spectroquant NOVA 30 from Merck (Darmstadt, Germany) and oxytop respirometers (WTW, Weiheim, Germany) were used for BOD analysis. All the other parameters were calculated according to the Standards Methods for the Examination of Water and Wastewater (APHA, 2012).

2.3. Statistical analysis of experimental data

Statistical treatment of data was carried out with the SPSS 24.0 statistic software (IBM Co. Ltd, USA). Pearson's correlation coefficient (r) was computed between different parameters in order to determine the extent to which values of both parameters were linearly correlated. The efficiency in microlitter and microplastic removal between different treatment stages was performed by paired Student's T-test, matching pairs samples prior and after a stage within the WWTP, and being no-difference made by the treatment stage the correct rejection of the null hypothesis. Besides, the fitting performance of analysis of variance (ANOVA) and general linear model for repeated measurements with estimated marginal means test were computed by means of F-test. All parameters were calculated at least at a 95% confidence level.

3. Results and discussion

3.1. General considerations

A total of 1163 ML particles were identified across all wastewater samples, by means of visual examination with the stereomicroscope and chemical composition by FTIR analysis. Most of them mainly consisted of soap components, including microparticles of calcium stearate, glycerin, stearic acid, lubricants and lipid mediators, reported to reduce irritation by aqueous detergents; silicon dioxide and silicates; cellulose; chipboard fragments and animal fur, besides microplastic particles. As recently reviewed by Li et al. (2018), it is very difficult to visually differentiate MP from other extracted organic and inorganic particles of similar size and shape, so additional approaches such as the spectroscopic approaches are required. These techniques also give an added value to the analysis, providing information about the polymer composition of MP after their visual preselection (Löder and Gerdts, 2015). Fig. 1 depicts images of both microplastic and non-plastic particles, in order to prove their similarity and their difficulty in differentiating them without an additional spectroscopic technique.

The average concentration of ML particles was 12.43 (±2.70) ML L^{-1}, 9.73 (±3.04) ML L^{-1}, 3.21 (±0.50) ML L^{-1}, and 1.23 (±0.15) ML L^{-1}, for GGR, PCL, BRT, and EFF, respectively, as depicted in Fig. 2. These outcomes proved a statistically significant removal of ML through the WWTP, accounting for a 90.1% between influent and effluent. ML particles proved to statistically significant decrease from PCL to BRT in a 67.0% (T-test = 2.257, $p = 0.042$). This finding is consistent with the role of a sedimentation tank or primary clarifier, used to remove both organic and inorganic materials, including grit, particulate matter, suspended solids and chemical flocs, among others (Murphy et al., 2016). Despite a statistically significant reduction of an additional 61.6% of ML from BRT to EFF (T-test = 4.741, $p = 0.000$), still about 10% of ML is emitted to the receiving waters, later reused for agriculture purposes. All sampling points contained a higher fraction of non-plastic than MP particles; i.e., 74.2% and 25.8% for GGR, 73.4% and 26.6% for PCL, 74.7% and 25.3% for EFF, except for BRT, with a 33.8% of non-plastic particles and 66.2% of MP.

MP comprised a 46.6% of total ML, with average concentrations also decreasing among wastewater treatment steps, as depicted in Fig. 2; i.e., 3.20 (±0.67) MP L^{-1}, 2.59 (±0.85) MP L^{-1}, 2.13 (±0.38) MP L^{-1}, and 0.31 (±0.06) MP L^{-1}, for GGR, PCL, BRT, and EFF, respectively, accounting for a removal of 90.3% and a maximum concentration of 13.04 MP L^{-1} in a GGR sample. The average value for GGR was low compared to the study reported by Murphy et al. (2016), with 15.70 MP L^{-1} in the influent, although the average concentration in the effluent was quite similar (0.25 MP L^{-1}). Simon et al. (2018) found much more particles per volume, with a median MP concentration of 54 particles L^{-1} in effluent wastewater, although they focused on the smaller particle size (10–500 μm). The highest decrease within our WWTP (85.4%) was between BRT and EFF (T-test = 4.947, $p = 0.000$) although, unlike ML, it was not significant between PCL and BRT (T-test = 0.395, $p = 0.700$). These results may indicate a clear influence of the biological reactor in the removal of MP, whether they are degraded by biological processes or transferred to the secondary sludge, and it will explain the high ratio between ML and MP in the effluent, compared to previous processes (Fig. 2). As reported by Mahon et al. (2017), the vast majority of MP becomes entrained in the sewage sludge. MP in the sludge can be transferred into the soil, because mixed sludge is used as a fertilizer in agriculture after anaerobic digestion.

Despite the important removal rate of the sewage plant, with an emission factor of about 9.7% higher than the average retention rate in Europe for WWTPs (Hann et al., 2018), an estimated 6.7×10^6 MP per day could be released with the final effluent; this is about 2.45×10^9 MP per year, similar to figures reported by Ziajahromi et al. (2017). The reduction in MP concentration was lower than that reported by Mintenig et al. (2017) in a study with 12 WWTPs in Germany, most of them provided with a tertiary treatment, settling processes in maturation ponds or final filtration.

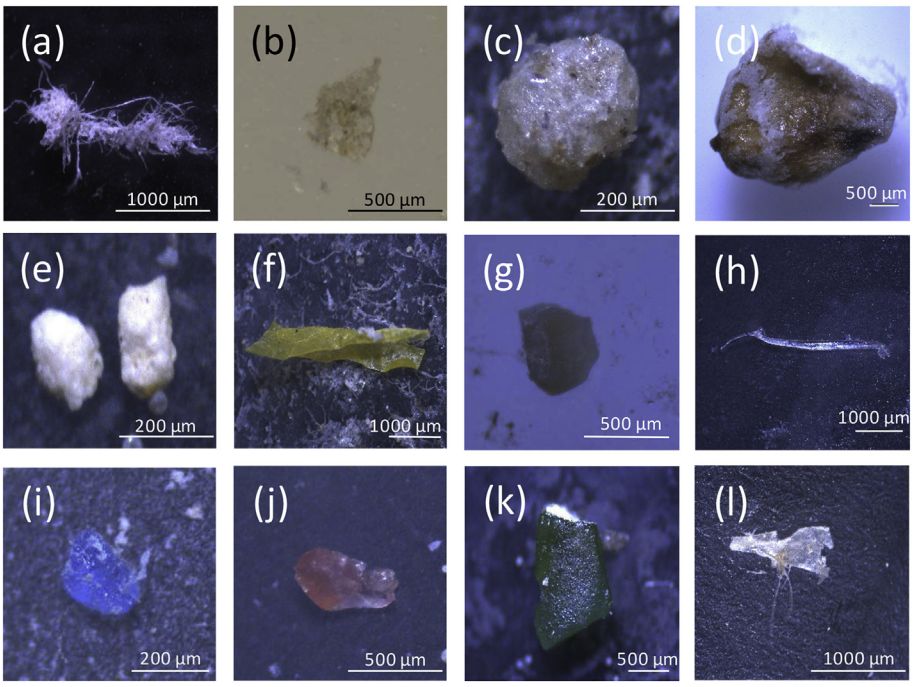

Fig. 1. Microlitter (a–e) and microplastics (f–l) in different stages of the WWTP identified by FT-IR: (a) cellulose (EFF/29th.Sep.2016); (b) silica dioxide (BRT/14th.Feb.2017); (c) soap (GGR/4th.Jul.2017); (d) chipboard (BRT/2nd.Oct.2017); (e) calcium carbonate (EFF/3rd.May.2017); (f) low density polyethylene (LDPE) (GGR/4th.Jul.2017); (g) ethylene propylene diene monomer rubber (EPDM) (BRT/13th.Mar.2017); (h) polyethylene terephthalate (PET) (BRT/22nd.Jan.2018); (i) low density polyethylene (LDPE) (GGR/5th.Jun.2017); (j) polypropylene (PP) (BRT/24th.Oct.2017); (k) polyethylene (PE) (EFF/17th.Nov.2017); (l) acrylate (ACRYL) (GGR/4th.Jul.2017).

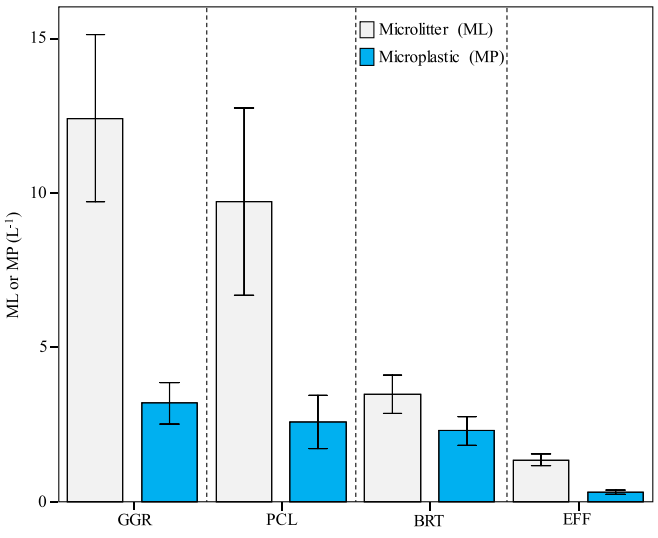

Fig. 2. Average concentrations of ML and MP in grit and grease removal (GGR) (n = 14), primary clarifier (PCL) (n = 14), biological reactor (BRT) (n = 18), and final effluent (EFF) (n = 19) (error bars represent standard error).

3.2. Analyses of microplastics in wastewater samples

3.2.1. Shape, color and size distribution

All the 1163 ML particles were analyzed by FTIR, although with different success rates (Fig. 3). As indicated by Talvitie et al. (2017), the biofilm contamination of microparticles together with extremely low sizes for some of them made different microparticles remained unrecognized. Also, the fact that many plastics are mixtures of polymers and copolymers makes them very hard to conclusively identify.

A total of 542 MP particles were identified by FTIR across all the examined samples. The five shapes detected were fragments (46.9%), films (34.0%), beads (11.5%), fibers (7.4%), and foam (0.2%). Fragments were the dominant type of MP in all seasons, comprising 50.6% in winter, 36.0% in spring, 59.6% in summer, and 45.1% in autumn, similar to data previously reported by Rodrigues et al. (2018) in water and sediments from a Portuguese river. Fig. 3(a) displays the evolution of these MP shapes through the WWTP, based on their concentrations. Films decreased from GGR (43.3%) to EFF (9.1%), conversely to fragment forms that increased during the sewage treatment, comprising a 60.5% of MP in the final EFF. Similar results were reported by Talvitie et al. (2017) for total ML in a WWTP. In any case, concentration of different shapes always decreased from GGR to EFF, as depicted in Fig. 4(b), fibers and beads being the less retained; i.e., 66.7% and 77.1% respectively, and directly released to the environment.

Most fibers were categorized as transparent (60.0%), as previously reported by Mintenig et al. (2017) in 12 WWTPs located in Germany, while most beads were classified as opaque (61.4%), a category selectively consumed by fish species as examined by Carpenter and Smith (1972). In most developed countries, the sewage infrastructure receives the effluent from domestic washing machines (Napper and Thompson, 2016) and, as reported by Browne et al. (2011), a single garment could produce >1900 fibers per wash, besides their lost due to pilling during wearing. In our study, fibers and fragments were the most prevalent microplastic forms within the effluent, as previously reported by Mason et al. (2016).

Fig. 4 depicts the proportion of different sizes of microplastics across the WWTP. Average MP size decreased from GGR 0.82 (±0.06) mm, to PCL 0.74 (±0.08) mm and then to BRT 0.63 (±0.03) mm, indicating that main wastewater treatment stages displayed a statistically significant removal of MP in their larger size fraction (*F*-test = 3.038, $p = 0.029$). However, these fractions displayed a

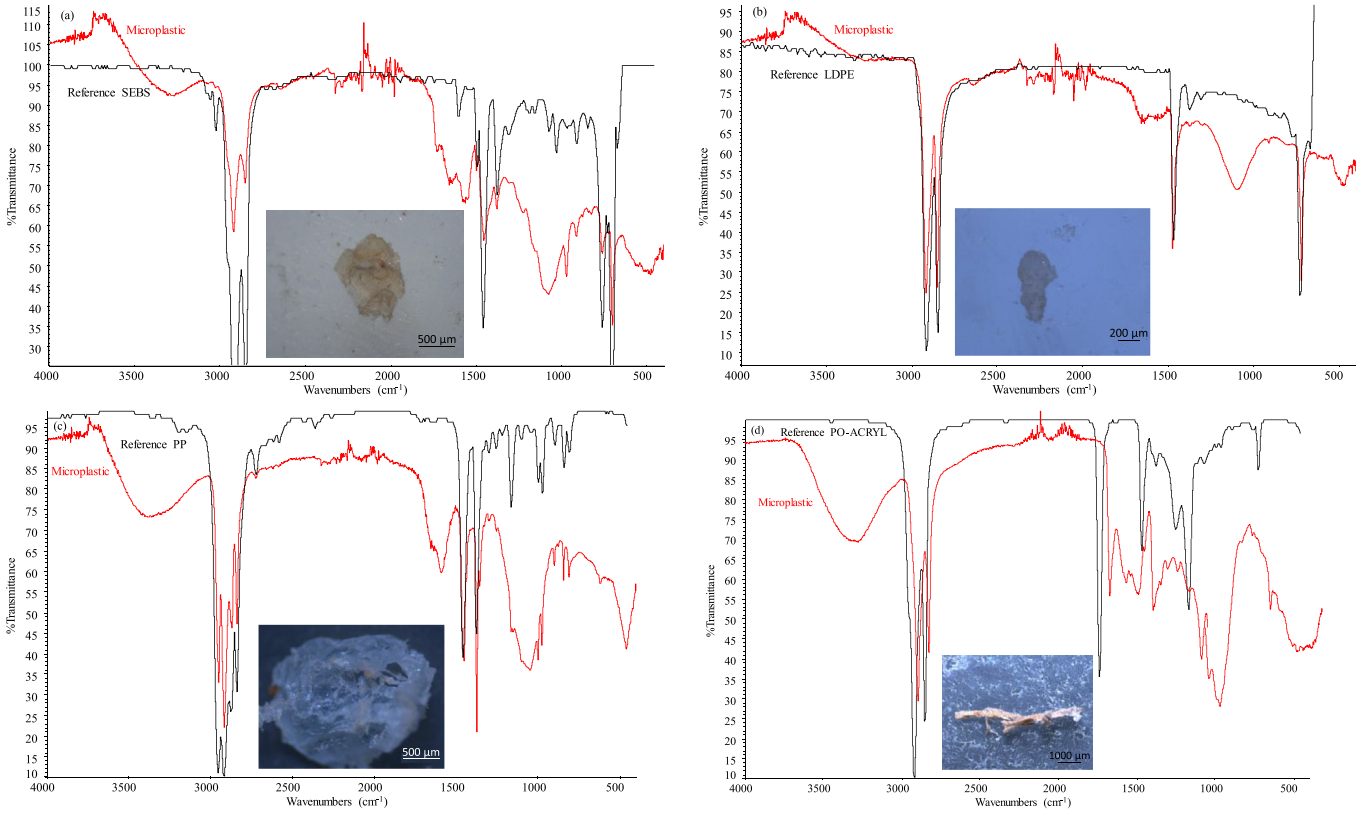

Fig. 3. Composition of microplastics in wastewater samples: (a) Poly(styrene:ethylene:butylene:styrene) BRT 13th.Mar.2017 78.81% match (Sprouse Scientific Systems Polymers by ATR Library); (b) Poly(ethylene) (low density) BRT 13th.Mar.2017 90.78% match (Sprouse Scientific Systems Polymers by ATR Library); (c) Polypropylene BRT 5th.Jun.2017 86.33% match (Hummel Polymer and Additives); (d) Poly(octadecyl acrylate) GGR 4th.Jul.2017 81.34% match (Hummel Polymer and Additives).

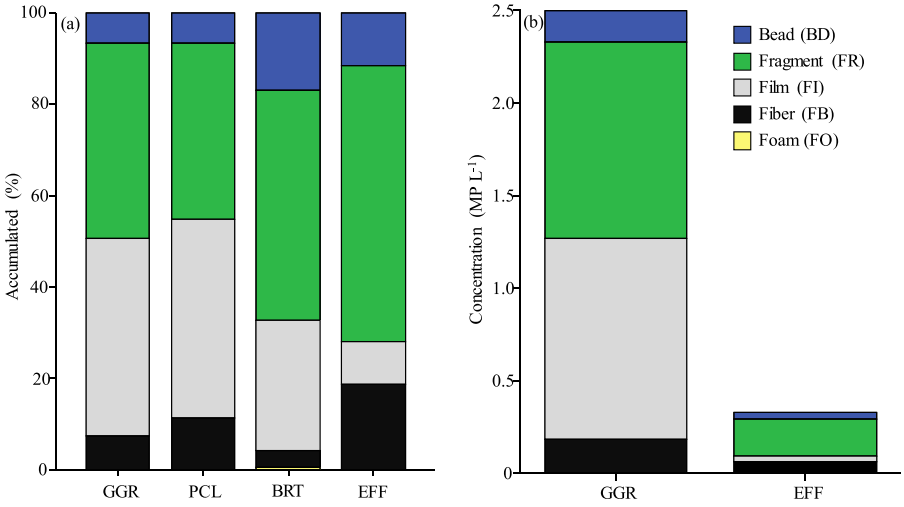

Fig. 4. Shape categories across the WWTP: (a) accumulated percentage based on MP concentration, and (b) concentration in the grit and grease removal (GGR) and final effluent (EFF).

statistically significant increase again in the EFF 0.83 (±0.14) mm compared to BRT (F-test = 4.880, $p = 0.028$), although it was not due to an increase in fibers concentration. In contrast to Browne et al. (2011), that reported the presence of only fibers in municipal effluent, we found different shapes of MP in the current study, as also proposed by Murphy et al. (2016).

As presented in Figs. 5 and 83.0% of recovered MP in all stages were smaller than 1 mm, a higher percentage than that reported by Lares et al. (2018) in a Finnish WWTP with seven samples; i.e., 64%. An average of 57.5% of MP were under 600 µm and, in all sampling points, most particles were of a size between 400 and 600 µm (35.2%). Similar to that described by Lares et al. (2018), the main size of MP in the final effluent was between 600 and 800 µm (26.5%).

The most common color for MP particles was beige (36.9%), followed by white (23.6%), black (7.8%), blue (7.0%), and green (3.9%). The identified fibers were mostly PET fibers (20.0%) in a blue color (27.5%), similar to those reported by Ziajahromi et al. (2017).

The most common polymer type for beads was low density polyethylene (LDPE) (57.1%), followed by high density polyethylene (HDPE) (19.1%), and polypropylene (PP) (17.5%). These results are in agreement with those proposed for microbeads included in commercial facial cleansers previously described by Bayo et al. (2017).

3.2.2. Polymers

A total of 17 polymer families were identified in different stages of the sewage plant, as presented in Table 1, with a 5.0% of unidentified microfibers that proved to be synthetic fibers due to their shiny surface, symmetrically round cross-section, uniformity of color and no cellular or other organic structure in their constitution (Talvitie et al., 2015; Mahon et al., 2017). Vandermeersch et al. (2015) propose the use of a hot needle to observe melting point and identify suspected plastic fiber. The highest average concentration was by far for LDPE ($2.83 \pm 0.47\ L^{-1}$), followed by HDPE ($0.94 \pm 0.41\ L^{-1}$), ACRYL (acrylate) ($0.83 \pm 0.30\ L^{-1}$), PP ($0.64 \pm 0.11\ L^{-1}$), PEP (polyethylene propylene) ($0.27 \pm 0.09\ L^{-1}$), PS (polystyrene) ($0.21 \pm 0.10\ L^{-1}$), BPL (biopolymer) and NYL (nylon) ($0.19 \pm 0.07\ L^{-1}$), PUR (polyurethane) ($0.14 \pm 0.08\ L^{-1}$), PET (polyethylene terephthalate) ($0.13 \pm 0.06\ L^{-1}$), MCR (methacrylate) ($0.11 \pm 0.07\ L^{-1}$), PTFE (Teflon) ($0.07 \pm 0.06\ L^{-1}$), MMF (melamine) ($0.04 \pm 0.02\ L^{-1}$), PES (polyester) and PVI (polyvinyl) ($0.03 \pm 0.01\ L^{-1}$), PIB (polyisobutylene) ($0.02 \pm 0.01\ L^{-1}$), and RBB (rubber) ($0.01 \pm 0.00\ L^{-1}$). Mintenig et al. (2017) detected MP comprised of 14 different polymers, polyethylene being the more common one in all size classes (40%). Our results match with the polymer resin types most demanded as raw material by European plastic converters, both in 2015 and 2016, and reported by Plastics Europe (2017).

In our study, the vast majority of isolated MP was LDPE (52.4%), 27.7% of them in a film form. There are at least two possible explanations for this result. The proximity of the WWTP to crops within agriculture greenhouses, mostly made up of ambient degraded LDPE (Serrano et al., 2004). Agriculture in a controlled environment is useful in deserts and temperate regions, where greenhouse structures are monitored with different input parameters to maintain the desired growing environment (Jensen, 2002). This covering material is dominant in the Mediterranean regions, being the most inexpensive plastic film (Papadakis et al., 2000).

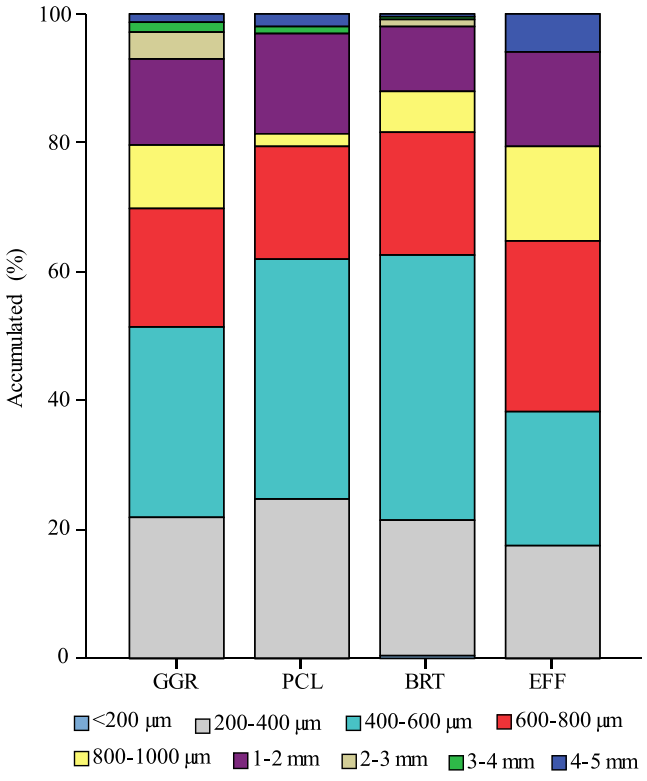

Fig. 5. Size categories at 4 different sampling points in the WWTP based on size classification from Spanish Environmental Ministry.

Fig. 6 depicts a comparison among a greenhouse sample, a microplastic from the primary clarifier and reference LDPE, in order to prove this fact. The other evidence that supports our hypothesis is the massive trade in single-use plastic bags, which free distribution has been recently banned in Spain. Future studies could more completely consider the consequences of this new legislation in our country, and its importance in the decrease of MP in wastewater, especially for LDPE.

Biodegradable polymers (BPL), represented by sebacic acid and caprolactone polymers, proved to significant decrease from GGR

Table 1
Polymer classes detected in GGR, PCL, BRT, and EFF of the WWTP, expressed as percentage (%) of isolated MP.

Polymer families	GGR (%)	PCL (%)	BRT (%)	EFF (%)	Total
Acrylate (ACRYL)	2.0	1.7	0.2	0.7	4.6
Biopolymer (BPL)	2.6	0.4	0.2	–	3.1
High-density polyethylene (HDPE)	1.3	2.8	4.4	0.6	9.0
Low-density polyethylene (LDPE)	13.8	9.2	27.3	2.0	52.4
Melamine (MUF)	–	0.2	0.6	0.2	0.9
Methacrylate (MCR)	0.6	–	0.4	0.4	1.3
Nylon (NYL)	0.7	0.6	0.7	0.4	2.4
Polyester (PES)	–	–	0.4	0.4	0.7
Polyethylene propylene (PEP)	1.8	–	0.7	0.4	3.0
Polyethylene terephthalate (PET)	0.4	–	0.9	0.6	1.8
Polyisobutylene (PIB)	–	–	0.4	–	0.4
Polypropylene (PP)	2.2	1.7	6.8	0.6	11.3
Polystyrene (PS)	–	0.7	0.6	0.2	1.5
Polyurethane (PUR)	0.2	0.2	0.7	–	1.1
Polyvinyl (PVI)	0.6	–	–	0.4	0.9
Rubber (RBB)	0.2	–	–	–	0.2
Teflon (PTFE)	–	0.2	–	0.2	0.4
Unidentified	1.3	2.0	0.6	1.1	5.0
Total	27.7	19.6	44.8	7.9	

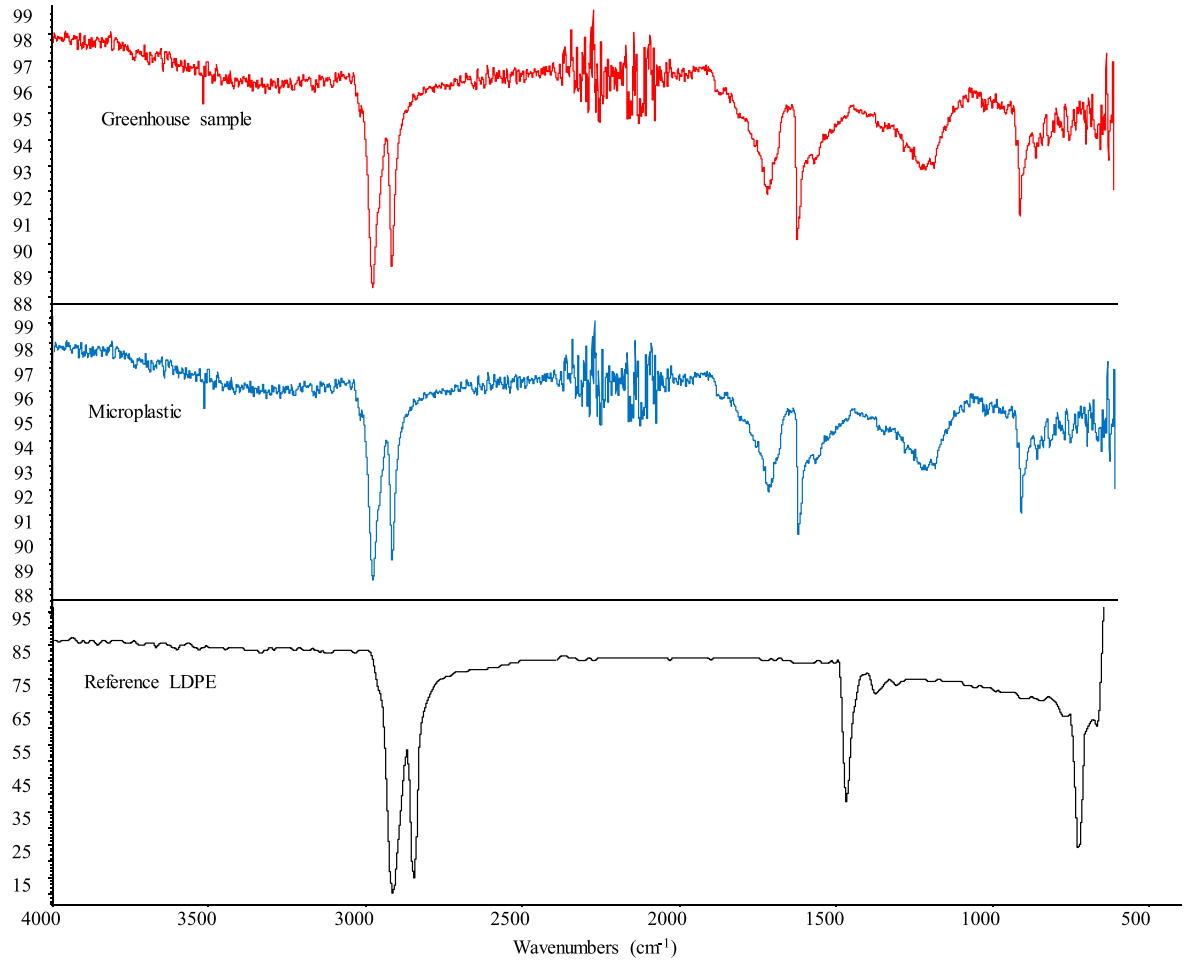

Fig. 6. FTIR for a greenhouse sample (LDPE with 75.54% match), Microplastic from primary clarifier 23rd.Jan.2019 (LDPE with 71.38% match), and reference LDPE (Sprouse Scientific Systems Polymers by ATR Library).

(0.21 ± 0.09 MP L^{-1}), down to PCL (0.02 ± 0.01 MP L^{-1}) and BRT (0.01 ± 0.01 MP L^{-1}), totally disappearing in the EFF. However, the removal was only statistically significant between GGR and PCL (*T*-test = 2.173, $p = 0.039$) and not between PCL and BRT (*T*-test = 0.589, $p = 0.561$). This can probably be explained by their own nature, undergoing quick and extensive biodegradation within the first treatment stages.

3.3. Relationships between microplastics and physicochemical parameters

Different physicochemical parameters in wastewater samples proved to be related to the presence and characteristics of MP. A statistically significant decrease on MP size was observed with low pH values ($r = 0.114$, $p = 0.010$), especially in BRT where the lowest pH data was processed; i.e., pH = 6.71, as well as the lowest average size (0.63 ± 0.03 mm). In our study, we also found a statistically significant decrease in PS concentrations with low pH ($r = 0.531$, $p = 0.004$). Strong mineral acids have proved to damage and/or destroy this polymer type (Cole et al., 2014), although it has also been proved that oxidation of polystyrene occurs in air when temperatures are elevated (Hurley et al., 2018). In any case, as previously reported in studies carried on microplastics in other environments (Karami et al., 2018), the synergistic effects of physical, chemical and biological factors in wastewater treatments could contribute to this fact.

Influent wastewater with high suspended solid concentrations were statistically related to a low MP burden ($r = -0.587$, $p = 0.001$). The aggregation of microplastics with particulate matter could increase their size by forming hetero-aggregates, because their reduced solubility and inertness (Besseling et al., 2017; Horton et al., 2017), leading to an increase in the sedimentation rate. In fact, MP larger than 1 mm were the predominant size in wastewater samples with a high suspended solids content, *versus* microplastics smaller than 1 mm (*F*-test = 4.740, $p = 0.000$). Similarly, wastewater samples with high COD values in the influent showed low average concentrations of PS ($r = -0.445$, $p = 0.020$) and PET ($r = -0.439$, $p = 0.022$). Both plastic polymers display surface energies of 42 and 34 mN m^{-1}, respectively, that could enhance the recruitment of fouling organisms from wastewater, as reported by Andrady (2011). All these results should raise awareness about the importance of increased interdisciplinary approaches for understanding MP dynamics in water systems. Nonetheless, an integrative understanding of physical, chemical and biological mechanisms driving MP evolution in the WWTP remains unclear.

The contribution of MP to the amount of oxidizable pollutants; i.e., COD, and to biological aerobic degradation; i.e., BOD was calculated, resulting in average values of 5.49 (± 1.04)·10^{-3} MP/mg COD and 5.22 (± 1.29)·10^{-3} MP/mg COD, for GGR and EFF, respectively, and 7.91 (± 1.49)·10^{-3} MP/mg BOD and 34.26 (± 7.73)·

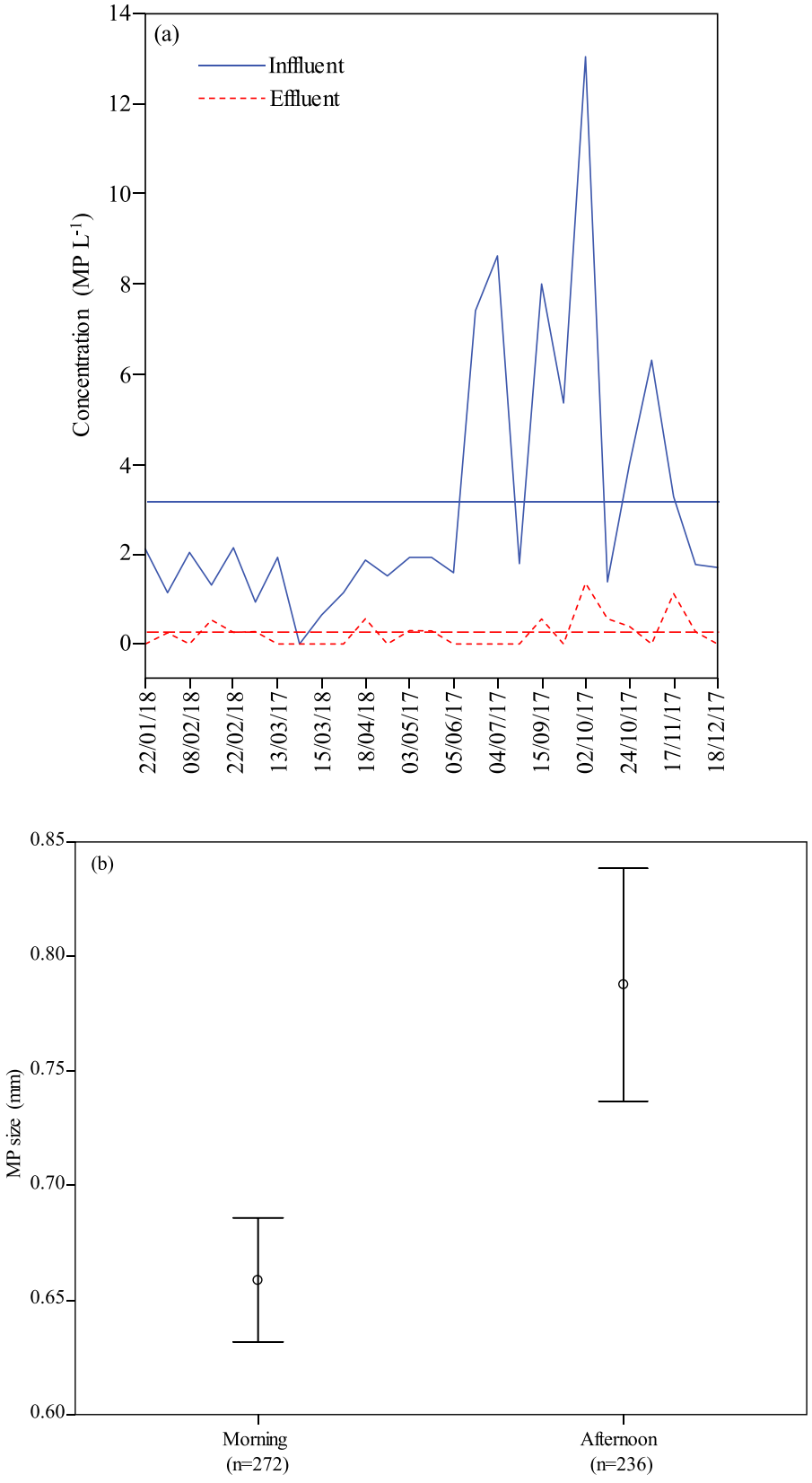

Fig. 7. (a) Temporal patterns of microplastic concentration in the influent and effluent of the WWTP by season, and (b) diurnal variation of MP size.

10^{-3} MP/mg BOD, for GGR and EFF, respectively. These data accounts for a roughly constant number of MP particles despite a COD reduction in the WWTP, from GGR (626.83 ± 19.06 mg L^{-1}) to EFF (52.64 ± 2.00 mg L^{-1}) (F-test = 3.581, $p = 0.014$), although an increase of MP particles in wastewater despite a statistically significant decrease in BOD from GGR (434.44 ± 23.65 mg L^{-1}) to EFF (8.42 ± 0.42 mg L^{-1}) (F-test = 3.883, $p = 0.009$). These factors must be taken into consideration when attempts are made to explain MP degradation as, even at their small size, they do not undergo ready biodegradation (Andrady, 2011).

There was also a statistically significant inverse correlation between MP concentration in the influent and available nutrients in the wastewater effluent, both for ammonium ions ($r = -0.558$, $p = 0.002$) and total nitrogen ($r = -0.548$, $p = 0.003$). Average ammonium concentrations proved to decrease in the effluent from wastewaters with low MP concentrations in the influent (0–1.38 MP L^{-1}) (26.17 ± 2.24 mg L^{-1} NH_4–N) to samples with the highest MP concentrations (8–13.0 MP L^{-1}) (14.40 ± 1.80 mg L^{-1} NH_4–N) (F-test = 3.807, $p = 0.017$). Similarly, average total nitrogen descended from wastewaters with low MP concentrations in the influent (0–1.38 MP L^{-1}) (36.29 ± 2.02 mg L^{-1} TN) to samples with the highest MP concentrations (8–13.0 MP L^{-1}) (22.00 ± 2.00 mg L^{-1} TN) (F-test = 4.074, $p = 0.013$). Cluzard et al. (2015) reported a greater recovery of ammonium in sediments with microplastics compared to those without them, and Green et al. (2016) discussed about an easy hydration of carbonyl groups in plastics, that can adsorb and hence sequester available nutrients. As reported by McCormick et al. (2014), MP from WWTP with adsorbed inorganic nutrients could serve as a biofilm support for bacterial growth, impacting with high trophic levels in the environment that prefer to feed on detritus.

3.4. Temporal patterns

As reported by Lares et al. (2018), most studies dealing with MP concentration in wastewater have been carried out within a period of a few days, not taking into account diurnal or seasonal variations; even their study did not cover the spring and summer seasons. Because of the extent of our monitoring campaign, we can deal with some of these questions.

Fig. 7 shows the seasonal variability of MP concentration for the influent and effluent of the WWTP [Fig. 7(a)] and diurnal variations due to MP size [Fig. 7(b)]. A statistically significant seasonal variation in the abundance of total MP could be observed for the influent, being higher for warm than for cold seasons (F-test = 5.660, $p = 0.004$). In order to estimate the seasonal variability of MP concentrations, a general linear model for repeated measurements with estimated marginal means test was applied. MP abundance showed statistical significant differences according to different seasons, being always higher for the summer than for the other three seasons (6.45 ± 1.58 MP L^{-1}) ($F = 6.819$, $p = 0.015$). However, when the general linear model was implemented for the effluent, no statistically significant differences were observed by seasons (F-test = 0.188, $p = 0.668$), indicating a similar and stable performance of the WWTP efficiency during the whole period, despite a higher MP load during the summer.

Increased concentration of MP in wastewater during the hot period could be explained by sun irradiance, leading to an easier evaporation of water, as previously reported for the evolution of ions and solids in the same WWTP (Bayo and López-Castellanos, 2016). Moreover, high temperatures are assumed to accelerate plastic degradation rate and, although heat weathering of mesoplastics and larger fragments of plastic litter mainly occurs in soil and sand, with lower specific heat than water (Andrady, 2011), still statistically significant differences could be observed for MP concentration in wastewater by temperature, with average values of 3.61 (±0.64) MP L^{-1} and 13.67 (±4.18) MP L^{-1}, when water temperature increased from 17.0 °C to 28.0 °C, respectively (F-test = 8.179, $p = 0.001$). Al-Salem (2009) demonstrated an enhanced degradation of polyethylene films when exposed to increased levels of sunlight intensity and higher temperatures in summer season. The second highest average concentration was observed during autumn (4.62 ± 1.36 MP L^{-1}), maybe due to heavy rainfall events during the rainiest season in our region (Ruiz Álvarez et al., 2017), inducing urban runoff of soil-retained MP into the sewage system as well as sewer overflows that have proven to be relevant for the entry of MP into the environment (Duis and Coors, 2016). Lee et al. (2013) also reported an increase of large microplastics (1–5 mm) after the rainy season, from 8205 particles m^{-2} in May to 27,606 particles m^{-2} during September. The runoff of microplastics induced by rainfall episodes within a dynamic environment, caused fragmentation to small sizes, being statistically significant smaller in autumn (0.65 ± 0.03 mm) than in any other season (F-test = 2.880, $p = 0.036$).

No statistically significant diurnal variation in the MP concentration was observed, although MP size proved to increase from the morning (0.66 ± 0.03 mm) to the afternoon (0.79 ± 0.05 mm) (F-test = 5.380, $p = 0.021$), always under 1 mm size.

4. Conclusions

The present study showed a statistically significant removal of ML (90.1%) and MP (90.3%) in the effluent of an urban wastewater treatment plant located in Cartagena, Southeast of Spain. Microplastics comprised a 46.6% of total ML, indicating the need for a specific differentiation technique for microplastic particles; i.e., FTIR spectroscopy. Fragments were the dominant type of MP in all seasons, proving to increase during the sewage treatment, conversely to film shapes. Average MP size decreased from GGR to PCL and BRT, although increased again in the final effluent, being between 400 and 600 μm the most frequent size interval (26.5%) in the final effluent. MP particles were made of 17 different polymer types, with the majority identified as LDPE (52.4%) in a film form (27.7%). The proximity of the sewage plant to agriculture greenhouses, together with still a massive trade of single-use bags and plastic packaging could be enough reasons for these results. MP larger than 1 mm were the predominant size in wastewater samples with a high suspended solid content. Some nutrients proved to decrease in the effluent, when wastewater samples were loaded with MP in the influent. The results indicated that, despite the significant differences in MP concentrations in the influent during the warm season, there were no significant differences in the effluent throughout the seasons, which suggested a stable performance of the WWTP.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2019.124593.

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